APPLICATION OF REGULARIZATION QUADRATIC PROGRAMMING TO MECHANICAL RELAXATION SPECTRUM CALCULATION

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The regularization quadratic programming approach to infer relaxation spectra
from experimental mechanical data was studied. It was found that the inferior
of the sum of the squared difference between the input data and the back cal-
culated values did not yield a satisfactory relaxation spectrum as the
regularization weighting parameter α was increased. A modification was made
to the function to be minimized so that all data points were equally weighted.

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Quadratic programming was then found to be sufficient to infer a reliable
relaxation spectrum if the input experimental data had a high degree of
accuracy. When the experimental data were not sufficiently accurate,
regularization over-regularized the high value region of the solution spec-
trum before it could improve the low value region. Decreasing the number
of sought after points in the solution spectrum can compensate for the
noise level in the input experimental data, and will allow inferring a
reliable relaxation spectrum from the experimental data.

FOREWORD

This report was prepared by the Polymer Branch, Non-metallic Materials Division. The work was initiated under Project No. 2419, "Nonmetallic and Composite Materials," Task No. 241904, Work Unit Directive 24190415, "Structural Resins." It was administered under the direction of the Air Force Materials Laboratory, Air Force Wright Aeronautical Laboratories, Air Force Systems Command, Wright-Patterson Air Force Base, Ohio, with Dr. F. E. Arnold as the AFML Project Scientist. Co-authors were Dr. C. Y.-C. Lee, Air Force Wright Aeronautical Laboratories, Materials Laboratory/Polymer Branch, and Dr. D. R. Wiff and Mr. V. G. Rodgers, University of Dayton Research Institute.

This report covers research conducted from May 1979 to May 1980.

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SECTION I

INTRODUCTION

The problem of inferring a mechanical relaxation spectrum in linear viscoelastic theory has been the subject of many papers (References 1-8). The experimentally obtained modulus functions are related to the relaxation spectrum through the equations (Reference 9)

$$E_\tau(t) = \int_{-\infty}^{\infty} H(\tau) \exp(-t/\tau) \, d \log \tau, \tag{1}$$

$$E'(\omega) = \int_{-\infty}^{\infty} H(\tau) \frac{\omega^2 \tau^2}{1 + \omega^2 \tau^2} \, d \log \tau, \tag{2}$$

and

$$E''(\omega) = \int_{-\infty}^{\infty} H(\tau) \frac{\omega \tau}{1 + \omega^2 \tau^2} \, d \log \tau, \tag{3}$$

where $E_\tau(t)$ is the stress relaxation modulus, $E'(\omega)$ and $E''(\omega)$ are the in-phase and out-of-phase components of the complex dynamic modulus, respectively, the $H(\tau)$ is the sought-after relaxation spectrum.

Any one of the above equations can be expressed in the general form of

$$\psi(x) = \int_a^b K(x,y) f(y) \, dy \tag{4}$$

or, in matrix notation

$$\underline{\psi} = \underline{\underline{K}} \, \underline{f} \tag{5}$$

where $\underline{\psi}$ is the measured experimental data of the problem resulting in a solution $\underline{f}$ and $\underline{\underline{K}}$ is the operator (the kernel in this case) that relates the two vectors. If $\underline{f}^\circ$ denotes the real solution, there should exist an "exact" set of $\{\psi^\circ(x)\}$ such that

$$\underline{\psi}^\circ = \underline{\underline{K}} \, \underline{f}^\circ \tag{6}$$

Experimentally, only an approximation $\underline{\tilde{\psi}}$ to $\underline{\psi}^\circ$ can be obtained.

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The task at hand is then to find a procedure to infer a solution $\tilde{\underline{f}}$ from $\tilde{\underline{\psi}}$, such that for a given $\tilde{\underline{\psi}}$, $\tilde{\underline{f}}$ is the best approximation of $\underline{f}°$. This means we must find an appropriate operator (or procedure) $\underline{B}$ (not necessary $\underline{K}^{-1}$), such that

$$\tilde{\underline{f}}^B = B\,\tilde{\underline{\psi}}\,. \tag{7}$$

One approach is to find a solution $\underline{f}^B$ which satisfies the condition that the function

$$N = (\tilde{\underline{\psi}} - \underline{\underline{K}}\,\tilde{\underline{f}}^B)^2 \tag{8}$$

to be a minimum.

If the problem is an ill-posed problem, the solution thus found may not be a good solution. Tikhonov (Reference 10) has found that in such instances, by adding a regularization term to the solution mapping procedure, $\underline{B}$, a different $\tilde{\underline{f}}*$ (the asterisk denotes the solution is obtained with regularization) can be obtained which is a better approximation to the real solution $\underline{f}°$ than $\tilde{\underline{f}}^B$. Using a scaling factor α, to control the magnitude of the regularization term contribution in the mapping procedure, $\underline{B}$, it was shown that there exists an optimum α which gives a minimum value to the square difference of $(\tilde{\underline{f}}* - \underline{f}°)^2$.

Wiff and Gehatia (Reference 11) have combined the regularization method and a quadratic programming algorithm to treat the ill-posed problem of determining a molecular weight distribution of polymers from sedimentation-diffusion equilibrium data obtained via ultracentrifuge experiments. Since in real situations a priori knowledge of $\underline{f}°$ is not available, the value of the optimum α is not known. However, it was found that the norm $||(\tilde{\underline{\psi}} - \underline{K}\,\tilde{\underline{f}}*)^2||$ will go through a minimum with increasing, α. This solution $\tilde{\underline{f}}'$ (the prime denoting the solution that gives the minimum norm) is a much better approximation of $\underline{f}°$ than $\tilde{\underline{f}}^B$. Using this approach the Regularization Quadratic Programming (RQP) method has been used successfully in the molecular weight distribution determination (Reference 12).

Wiff (Reference 1) had applied the RQP method to the problem of inferring relaxation spectra from modulus data and the preliminary results were promising. This work is a more in-depth study of the problem. It is found that regularization will give a minimum norm of $||(\tilde{\psi} - \underline{\underline{K}}\,\tilde{\underline{f}}*)^2||$ only if different integration routines are used in the forward mapping operator B and the back calculation operator $\underline{\underline{K}}$. And because of the nature of the relaxation spectrum, regularization is not suitable for this problem. A modification to the expression to be minimized (Equation 7) was made. By adjusting the number of output points, a reasonable approximation of a known relaxation spectrum has been obtained.

SECTION II
REGULARIZATION QUADRATIC PROGRAMMING (RQP)

Quadratic programming can be used to find the minimum of the expression N (Equation 8). The algorithm (Reference 13) will find a solution $\underline{x}$ that minimizes the expression

$$M = \underline{x}\ \underline{\underline{D}}\ \underline{x} - 2\ \underline{\underline{E}}\ \underline{x} \qquad (9)$$

if D and E are given. Expanding Equation (9), one can show (with daggers denoting adjoints)

$$N = \underline{\tilde{f}}^{\dagger}\ \underline{\underline{K}}^{\dagger}\ \underline{\underline{K}}\ \underline{\tilde{f}} - 2\ \underline{\tilde{\psi}}^{\dagger}\ \underline{\underline{K}}\ \underline{\tilde{f}} + \underline{\tilde{\psi}}^{\dagger}\ \underline{\tilde{\psi}} . \qquad (10)$$

Comparing with Equation (9) and dropping the last term because it is a constant, it is obvious that

$$\underline{\underline{D}} = \underline{\underline{K}}^{\dagger}\ \underline{\underline{K}} ,$$

$$D_{ij} = \sum_{\ell} K_{\ell i}\ K_{\ell j} ; \qquad (11a)$$

and

$$\underline{\underline{E}} = \underline{\tilde{\psi}}\ \underline{\underline{K}}$$

$$E_i = \sum_{\ell} \tilde{\psi}_{\ell}\ K_{\ell i} . \qquad (11b)$$

The regularization term is

$$\Omega^{(n)}\ [f(y)] = \int_a^b \left[\frac{d^n f(y)}{dy^n}\right]^2\ dy \qquad (12)$$

By minimizing this term plus the functional in Equation (10) the solution $\underline{\tilde{f}}*$ is smoothed to remove noises which were assumed present due to the ill-posedness of the problem. Expressing the derivative in terms of binomial expansion:

$$\frac{d^n f(y)}{dy^n} = \frac{1}{h^n} \sum_{k=0}^{n} \binom{n}{k} (-1)^k\ f_{j-p+k} \qquad (13)$$

where h is the constant increment in y, $\binom{n}{k}$ is the binomial coefficient, and p=n for n=odd and p=n-1 for n=even. Substituting

Equation (13) in the matrix form of Equation (12), the regularization form is then

$$\Omega^{(n)}[f] = \frac{1}{h^{n-1}} \sum_{g=1}^{j} \sum_{r=1}^{j} \sum_{k=0}^{n} \sum_{l=0}^{n} \binom{n}{k}\binom{n}{l} (-1)^{1+k} f_{g-p+k} f_{r-p+l}, \quad (14)$$

which in matrix notation will be

$$\Omega^{(n)}[f] = \underline{f}' \underline{\underline{\Lambda}}^{(n)} \underline{f} \quad (15)$$

The RQP method then uses quadratic programming to minimize the function

$$M = N + \alpha\Omega \quad (16)$$

where N and Ω are defined in Equations (10) and (15), respectively, and α is a scaling factor that controls the relative contributions of the two terms to the total function M. With α too small, the ill-posedness of M was postulated to result in too much noise in the solution $\underline{\tilde{f}}^*$; but when α was too large, the solution became over-smoothed and deviated from the sought-after function $\underline{f}°$.

The scheme (I) used in Reference [1] varies α to obtain a set of $\underline{\tilde{f}}^*$, then the norms

$$||(\underline{\tilde{\psi}} - \underline{\underline{K}} \, \underline{\tilde{f}}^*)^2|| \quad (17)$$

were calculated, and the function $\underline{\tilde{f}}'$ that gives the inferior norm value was used as the best approximation of $\underline{f}°$. The numerical integration used in the term N (Equation 16) was the Trapezoidal Rule, while the Simpson's Rule was used in calculating the norm (Equation 17).

Modifications to this program have been made during this work. The new scheme (II) has two major differences from scheme (I). Firstly, the expression N is changed to reflect the relative difference, rather than the linear difference of the data points.

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The new expression is

$$N' = \left(\frac{\underline{\underline{K}}\,\tilde{\underline{f}}}{\tilde{\underline{\psi}}} - 1\right)^2$$

$$= \frac{\tilde{\underline{f}}^\dagger\,\underline{\underline{K}}^\dagger\,\underline{\underline{K}}\,\tilde{\underline{f}}}{\tilde{\underline{\psi}}\,\tilde{\underline{\psi}}} - 2\,\frac{\underline{\underline{K}}\,\tilde{\underline{f}}}{\tilde{\underline{\psi}}} + 1 \qquad (18)$$

Comparing with Equation (10),

$$D_{ij} = \sum_\ell \left(\frac{K_{\ell i}}{\tilde{\psi}_\ell}\right)\left(\frac{K_{\ell j}}{\tilde{\psi}_\ell}\right) \qquad (19a)$$

and

$$E_i = \sum_\ell \frac{K_{\ell i}}{\tilde{\psi}_\ell} \qquad (19b)$$

Secondly, the integration routine in the calculation of norm is changed to Trapezoidal Rule, to be consistent with the routine used in Equation (18).

In both schemes, the domain of the solution has to be specified. The range of τ for the relaxation spectrum should be large enough to cover the input data range. If t_1 and t_2 (ω_1 and ω_2 for storage and loss modulus data) are the lower and upper limit of the input data, then $\tau_1 < t_1$ and $\tau_2 > t_2$ (or $\tau_1 < 1/\omega_1$ and $\tau_2 > 1/\omega_2$). This is because the kernel for each value of $E(t)$ covers a range of τ. For $E(t_1)$, contributions from $H(\tau)$ for $\tau < t_1$ are present. By setting a limit of τ_1 the solution form is forced to have $H(\tau) = 0$ for $\tau < \tau_1$. If no $H(\tau)$ value is allowed for $\tau < t_1$, errors will be forced into the region between τ_1 and τ_2 to account for the relaxation effect in $E(t_1)$. For the same reasoning, the $H(\tau)$ values obtained near both τ limits should be regarded with caution. Unlike other experimental data, there is no guarantee that the mechanical modulus data is complete, and fully account for the changes reflected by $H(\tau)$. Often, instrument sensitivity determines the range of the input data, or in a master curve obtained near the glass transition region, the limit in the short time region is where the experimenter believes that the assumptions underlying the time-temperature superposition principle are no longer valid.

SECTION III
RESULTS AND DISCUSSION

The published experimental data of Tobolsky and Catsiff [Reference 14] on polyisobutylene were used to check the capability of scheme (I) to determine relaxation spectrum from mechanical data. Figure 1(a-c) shows the $H(\tau)$, $(\tilde{\underline{f}}')$ results generated from the stress-relaxation, storage and loss moduli, respectively, and all of which contain unacceptable levels of noise in the solution forms.

Figure 1(d) is the comparison between the initial stress-relaxation data and the back-calculated modulus values using the computer generated $H(\tau)$. It is obvious that the high magnitude data fit better than those of lower magnitude, which is not surprising because the initial data vary in magnitude over three decades (10^{10} to 10^7 dynes/cm^2). When the computer algorithm minimizes the sum in Equation (10), the terms from the large magnitude data will dominate the minimization process. By changing the sum to be minimized in scheme II to Equation (18), which considers the relative deviation of each data point instead of the absolute magnitude of deviations, all data points are weighted equally in the process.

In the original RQP scheme, without regularization, the quadratic programming algorithm is supposed to yield a solution $\tilde{\underline{f}}$, out of all possible solutions in solution space F, which gives a minimum norm $||(\tilde{\psi} - \underline{\underline{K}} \ \tilde{f})^2||$. It seems contradictory to say that with regularization, another solution in space F can be found which gives an even lower value of the norm. It is noted also that in all cases using the newly defined norm (Equation 18) the $\underline{f}'$ solution contains only slight deviations from the $\tilde{\underline{f}}$.

Further investigation of the computer routine showed the integration form assumed in Equations (11a and b) is different from the form used in the back calculation (Equation 17). The N values at the low α values not only contain the intrinsic sum
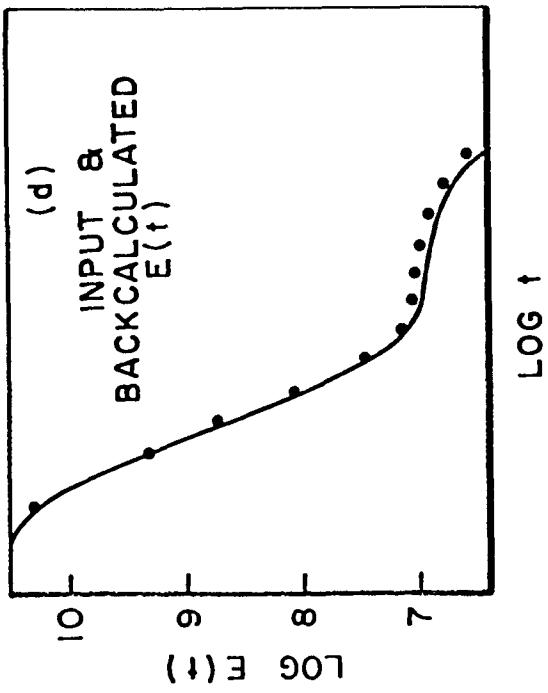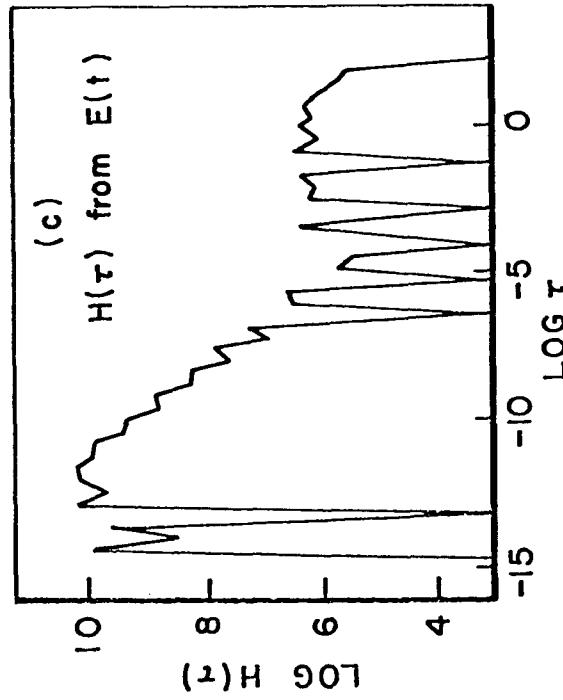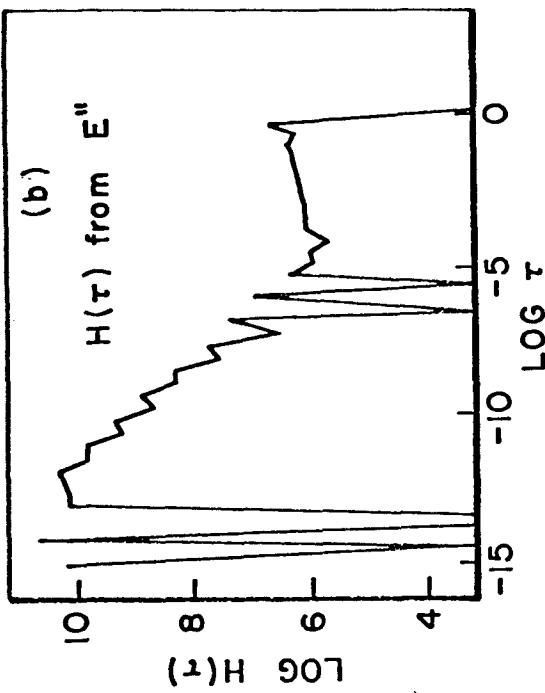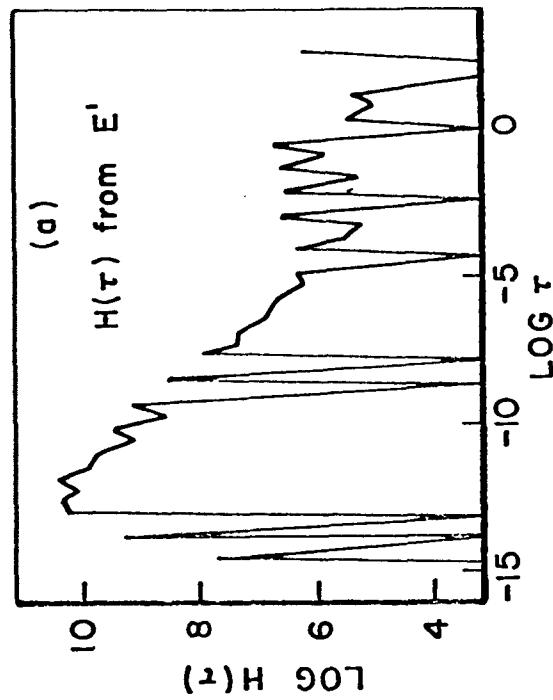
Figure 1. (a-c) The calculated $H(\tau)$ results of Scheme I using the data of Tobolsky and Catsiff on storage modulus, loss modulus, and stress relaxation modulus. (d) The input data of stress relaxation modulus is compared with the back-calculated values.

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of least squares of the problem, but also a small but discernible part due to the difference in the integration forms assumed in the forward and backward mapping. The back calculation integration uses the Simpson's Rule, while the forward mapping uses the Trapezoidal Rule. As the value of α increases, the part in N attributable to the different integration forms will decrease. With further increase in α, the regularization begins to over-smooth the solution form, such that N increases again, producing an artificial minimum in N.

In scheme II, the backward mapping integration routine was changed to the same form as that of the forward mapping. Figure 2 shows the N values as a function of α in schemes I and II. In scheme I, a minimum is observed, but the effect of regularization on the solution form at this value of α is minimal comparing with the solution form obtained at $\alpha=0$. In scheme II, as expected, N is an increasing function with increasing α.

Using the relaxation spectrum given by Tobolsky and Catsiff [Reference 14], the three sets of data (in log form) were generated using Equations (1-3). All data contain up to eight significant figures. Using scheme II, all sets gave very good approximations of the original relaxation spectrum (Figure 3) without regularization. The number of significant figures in the data sets were gradually reduced, and the unregularized solution forms in all cases gradually show discernible noise levels. Finally, a "white spectrum" of noise, is added to the two significant figure data of log modulus. The noise is scaled as such that the data will contain 2% noise. The unregularized solution form is shown in Figure 4a. This clearly demonstrates that quadratic programmingalone using the reduced norm will give good approximations of the relaxation spectrum, providing that the initial data contain sufficient degree of accuracy.
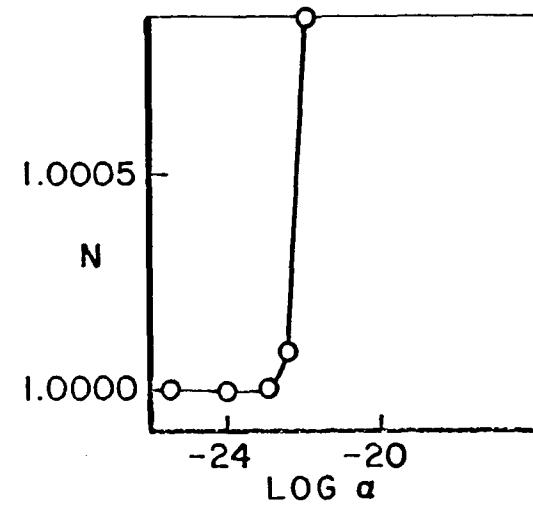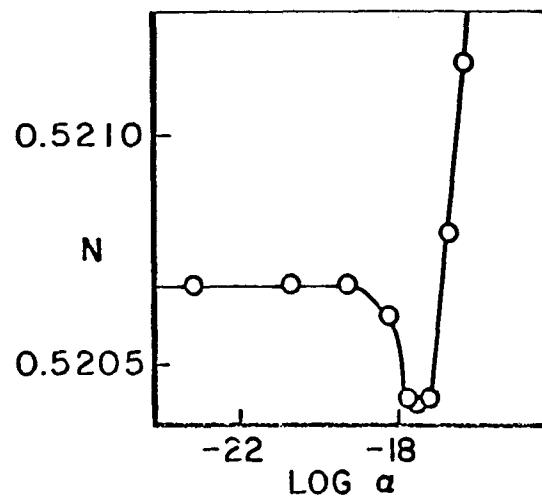
Figure 2. The N values as a function of α. The left is calculated with Scheme I, and the right with Scheme II.
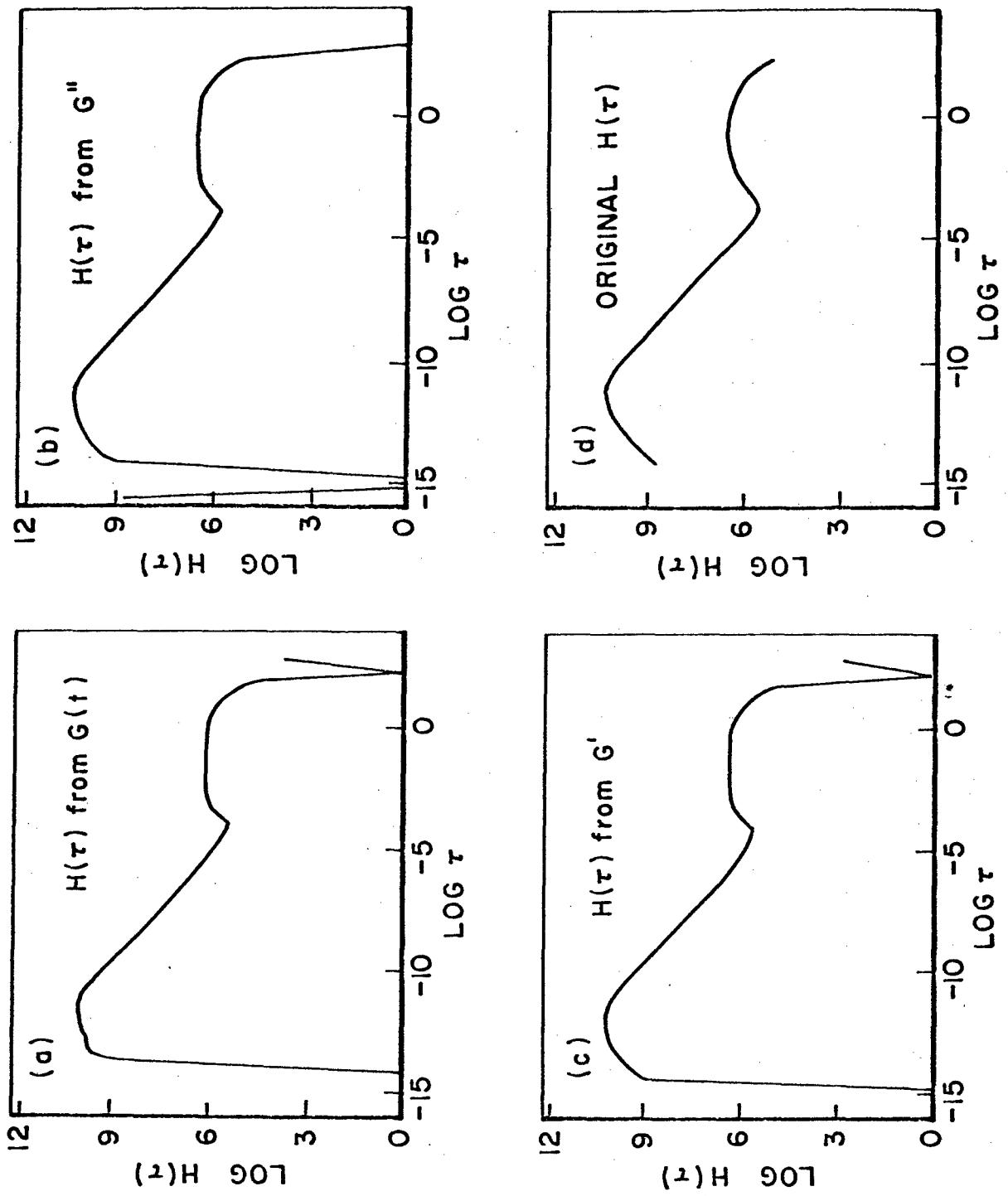
Figure 3. The H(τ) results from Scheme II with eight significant figures, data calculated from the assumed solution form displayed in (d). No regularization was used.
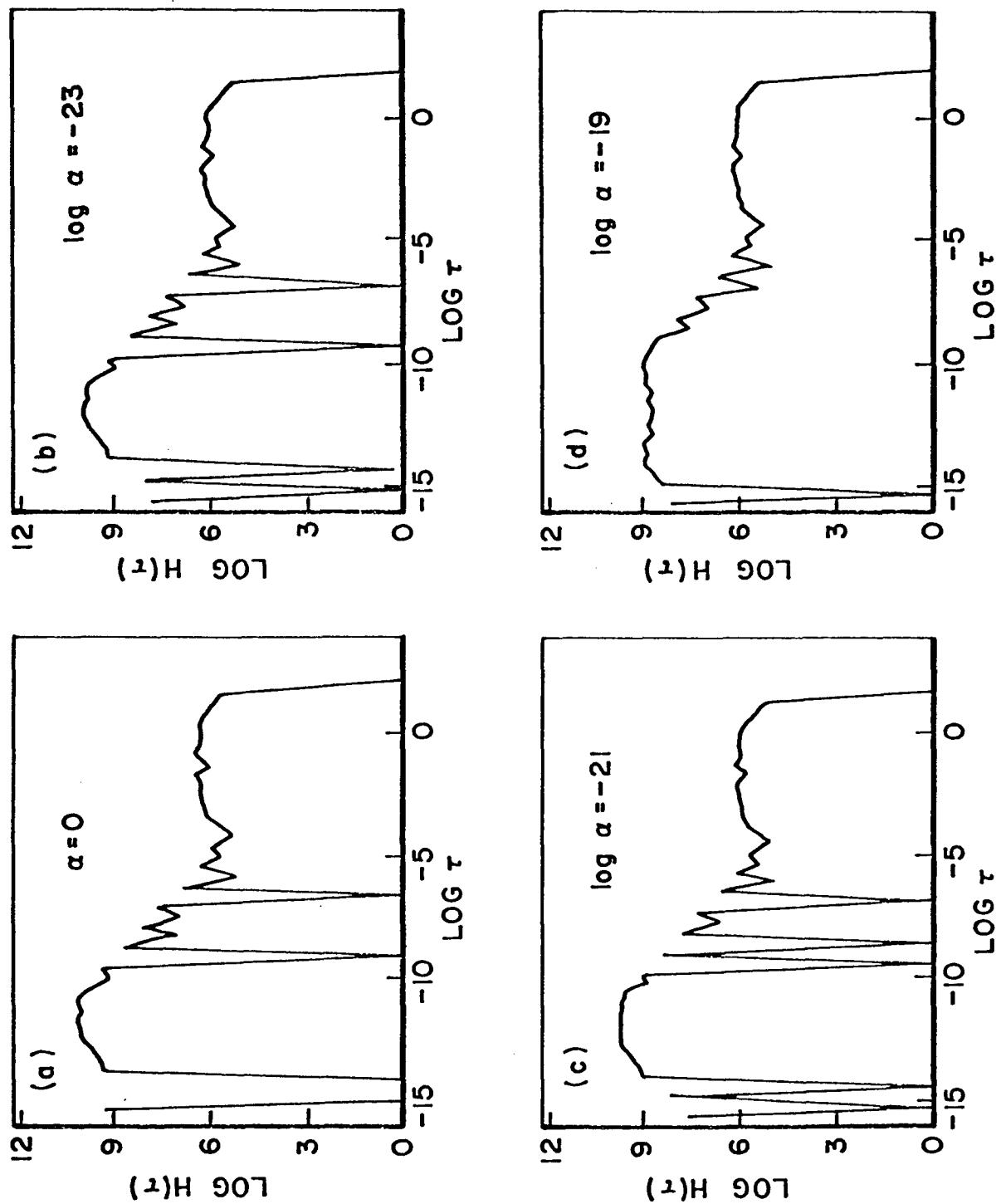
Figure 4. The $H(\tau)$ results using the calculated stress relaxation data containing 2% noise. The values of the regularization weighting parameter α used were (a) 0.0, (b) 1.0×10^{-23}, (c) 1.0×10^{-21}, and 1.0×10^{-19}.

The regularization functional was used in an attempt to remove the noise on the solution form obtained from the data with the 2% noise. The scaling factor α which controls the relative contribution of the regularization was gradually increased. At log α = -19 (Figure 4) the big valleys in the middle of the τ ranges were diminished, but the high values at the low τ range also suffered from over-regularization. Several n-values (Equation 12) were tested and all gave similar results. This is not really surprising considering the way the regularization term is written. The equation is regularizing the linear form of the solution, so the "real" slopes in the high magnitude region are larger or comparable to that of the noise in the low region. Regularization of the low region necessarily has to over-regularize the high magnitude region. One obvious alternative is to regularize only the region that contains noise, but then a-priori knowledge of what the solution form should be is required.

Another alternative is to rewrite the regularization term such that the function to be regularized is log $H(\tau)$, such that the "real" slope values are reduced. But the term will still have to be expressed in the linear form, so that it can be added to the term N for the quadratic programming routine. The way to accomplish that is not immediately obvious. But even if this can be accomplished, one is still faced with the problem of knowing what α value will best remove the noises in the solution form, yet not over-regularize it.

The apparent success of the regularization routine to substantially improve the solution form of the molecular weight distribution (MWD) determination may be due to two factors: (1) the problem is more "ill-posed"; (2) and the solution $\underline{f}^\circ$ takes on only a small range of values. Not every equation in the form of Equation (4) is "ill-posed"; it depends on the nature of the kernel. Obviously if the kernel is a delta function, the problem would be extremely well posed. For the molecular weight distribution determination problem, the problem is so ill-posed

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that the unregularized solution is dominated by noise; so much so that it almost resembles a random pattern [Reference 11,12]. It is possible that the two integration forms employed in the forward and backward mapping procedures produce a much bigger effect on the back-calculated value of N. After regularization has substantially improved the solution form, the effect from the two routines is minimized. So the function outputted as $\tilde{\underline{f}}'$ is a much better approximation of $\underline{f}^\circ$ than $\tilde{\underline{f}}$. Also in the MWD determination, the solution is in units of mole-fractions, and the numbers do not vary over several orders of magnitude like the relaxation spectrum. If one chooses to display the relaxation spectrum in a linear scale rather than the log scale, regularization will appear to work, too. In light of this discussion, it seems interesting to reinvestigate the MWD determination with the two integration routines being the same, but unfortunately that is beyond the scope of the present work.

Keeping the number of initial data points (85) constant, the number of spectrum computed points was varied. With the 2% noise data, the noise on the solution form decreases with decreasing number of spectrum points (Figure 5); and the solution obtained with 33 spectrum points became a reasonable approximation of the original relaxation spectrum. This means that by keeping the output/input data-point ratio low, the quadratic programming can compensate for the noise level in the initial input data. This is verified by increasing the number of output points with the set of data without noise. The acceptable solution form obtained previously became noisy.

Figure 6b shows the solution form obtained by scheme II using Tobolsky's experimental data. The number of spectrum points were decreased to 31 points (with 85 points of input modulus data). Figure 6a shows the solution form obtained with scheme I under identical conditions. Obviously, by decreasing the number of output points both schemes will decrease the noise level in the solution form (compare with Figure 1). Because scheme II is
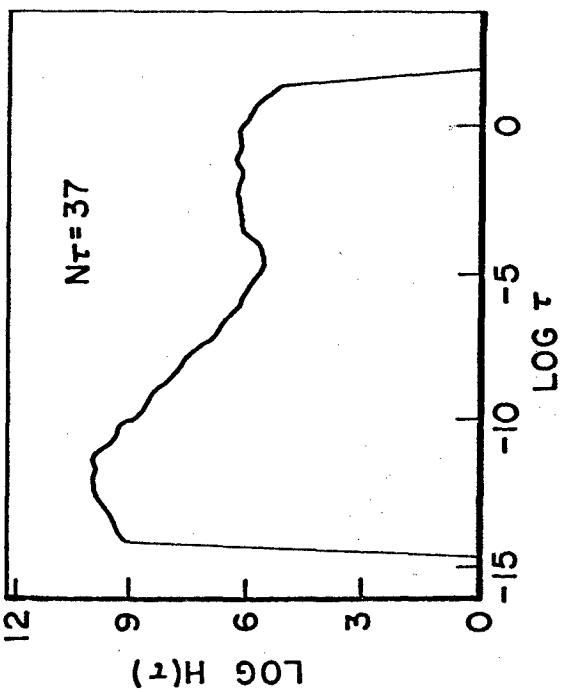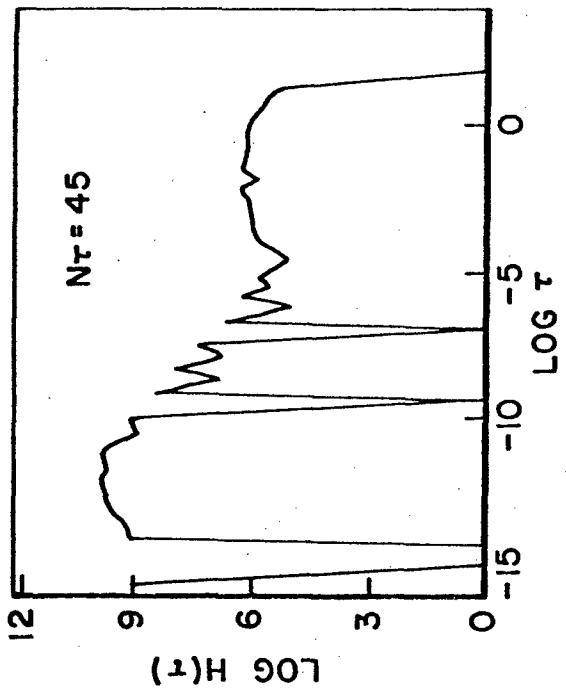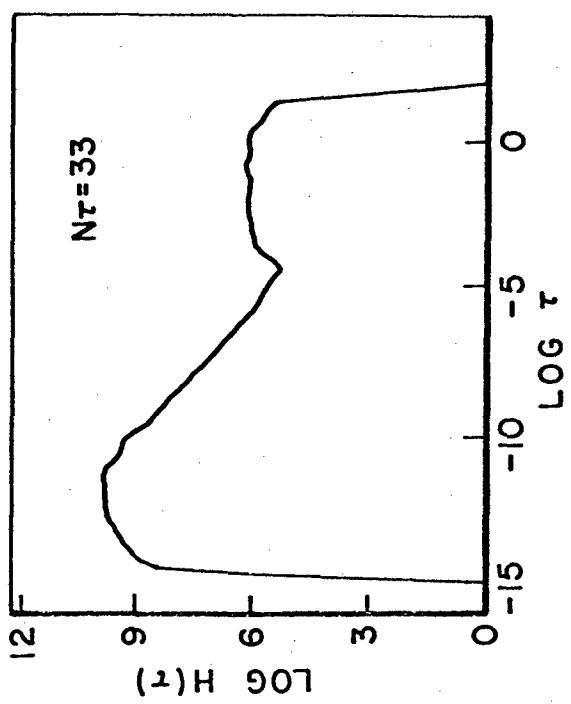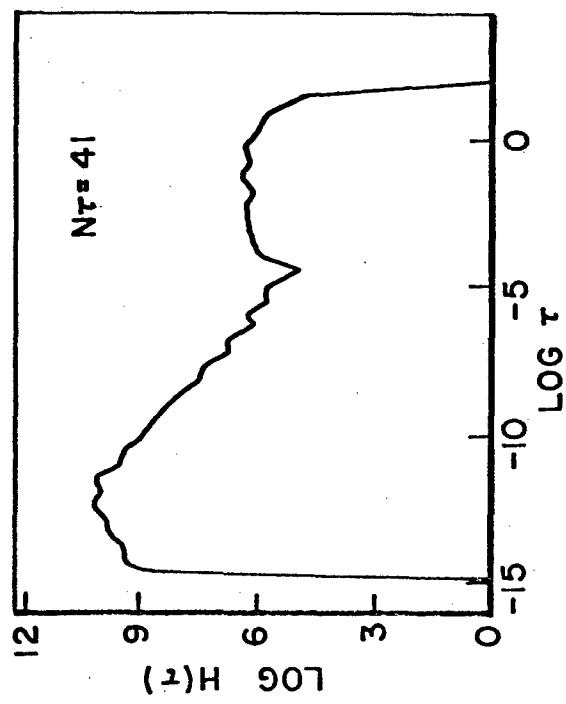
Figure 5. The H(τ) results using calculated stress relaxation data containing 2% noise. No regularization was used. The number of output points were displayed as Nτ.
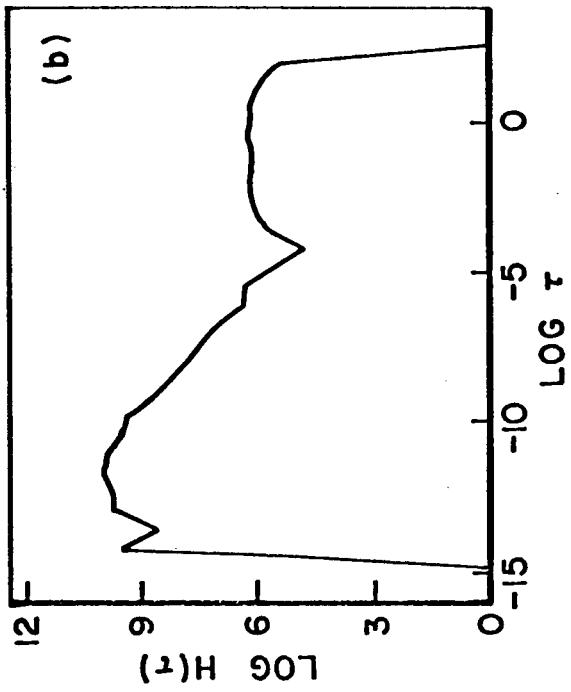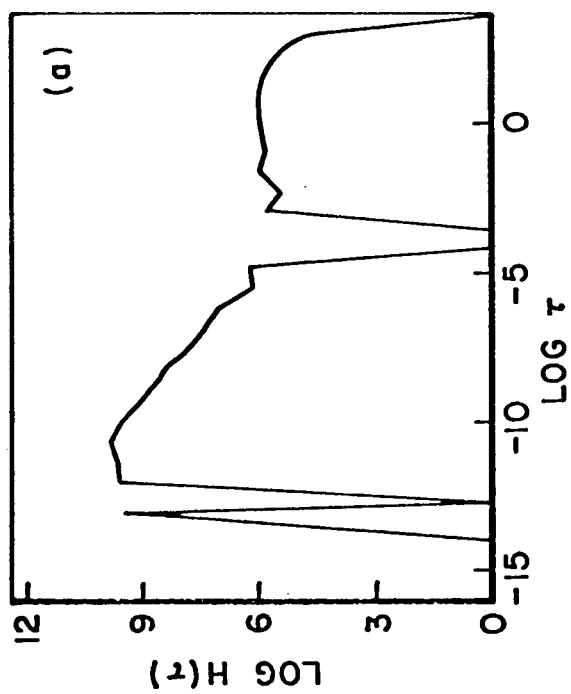
Figure 6. The $H(\tau)$ results using the stress relaxation data of Tobolsky and Catsiff. The number of output points were 33 and no regularization was used. The results from Scheme I are displayed in (a) and from Scheme II in (b).

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weighting each data point equally in its function to be minimized, under identical conditions, scheme II gives better solution than scheme I. Or, it can be said that scheme II can tolerate higher degree of noise in the input data.

Since the ratio of the output/input points is a determining factor in obtaining reasonable solution form, it would be interesting for future works to see if better solutions can be obtained from a given set of data by artificially increasing the number of input points through interpolation.

Because of the similar shape of the kernels between the storage and stress-relaxation modulus equations, the two sets of data behave very similarly in the quadratic programming routine. For the loss modulus data, the kernel does not overlap such a wide region of τ, that for a given noise level, the output/input ratio needed to give a reasonable relaxation spectrum is higher than a comparable storage or stress relaxation data. Since experimentally, storage and loss moduli are usually obtained with the same measurement, it would be interesting for future works to combine both sets of data in the quadratic programming routine to see if that would have the same effect as increasing the number of input data points.

SECTION IV
CONCLUSION

The regularization quadratic programming approach to infer relaxation spectra from experimental mechanical data was studied. It was found that the expression $\Sigma(\tilde{\psi}_i - K(f_i))$ will not give a minimum as the regularization weighting parameter α is increased. A modification was made to the function to be minimized so that all data points will be weighted equally in the program. Quadratic programming is found to be sufficient to infer relaxation spectra if the input data have a high degree of accuracy. When the data are not sufficiently accurate, regularization will over-regularize the high value region before it can improve on the low value region because of the nature of the relaxation spectrum. Lowering the number of output points can compensate for the noise level in the input data, and will allow inferring relaxation spectra from experimental data.

SECTION V
ACKNOWLEDGMENT

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SECTION VI

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